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Nucleation Controlled by Non-Fickian Fractional Diffusion

Vyacheslav Svetukhin

Scientific-Manufacturing Complex "Technological Centre", 124498 Moscow, Russia; svetukhin@mail.ru

Abstract: Kinetic models of aggregation and dissolution of clusters in disordered heterogeneous materials based on subdiffusive equations containing fractional derivatives are studied. Using the generalized fractional Fick law and fractional Fokker–Planck equation for impurity diffusion with localization, we consider modifications of the classical models of Ham, Aaron–Kotler, and Lifshitz–Slezov for nucleation and decomposition of solid solutions. The asymptotic time dependencies of supersaturation degree, average cluster size, and other characteristics at the stages of subdiffusion-limited nucleation and coalescence are calculated and analyzed.

Keywords: anomalous diffusion; fractional equation; cluster; nucleation; diffusion-limited process

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1. Introduction

In structural alloys of nuclear technology under different physical conditions (heating, irradiation, deformation, etc.), various kinds of defects are formed, which can lead to a significant change in the diffusion conditions of alloy components. The presence of dislocations, grain boundaries, and other material inhomogeneities can lead to noticeable deviations of particle diffusion from the classical behavior determined on the basis of Fick's law. A striking example of anomalous kinetics in solids is dispersive transport [1,2], which is characterized by the universality of properties and is observed in many disordered materials (see, e.g., [2–6]). This type of anomalous transport does not obey Gaussian statistics and is not described by Fick's law and the classical Fokker–Planck equation. In works [4–22], a fractional-differential approach is accomplished, which, in contrast to others, allows describing the normal and dispersive transport within a unified formalism. Within the fractional approach, many results of the diffusional decomposition theory of supersaturated solid solutions can be generalized to the case of dispersive self-similar transport in disordered solids. Some results in this direction are presented in [23,24].

The developed models can be useful for describing nucleation processes in polycrystalline materials. In [25,26], the formulation of grain-boundary diffusion in terms of the fractional calculus is proposed. The direct connection of this model with the well-known Fischer model and its modifications is demonstrated. The obtained solutions of fractional diffusion equations agree with the classical results of Whipple and Suzuoka. This formulation has allowed us in [25] to substantiate a new mechanism for the subdiffusion-assisted precipitation and to find a relationship between model constants and physical parameters of the material.

In [27,28], a new model is developed for hopping transport in disordered nanostructured materials, which takes into account the percolation structure of the sample and energy distribution of localized states. The model agrees with the results of Monte Carlo simulation. The combined effect of structural and energy disorder is taken into account in [27–29] within the framework of multiple trapping and hopping transport on comb structures [30,31].

In [32], the non-Markovian stochastic Liouville equation is used to study memory effects in bimolecular subdiffusion-assisted reactions. The non-Fickian type diffusion is found to slow down the kinetics of both geminate and bulk reactions. In [33], fractional kinetics of geminate reactions controlled by the subdiffusive relative motion of reacting

particles are studied in detail. These results are mainly applied to the description of recombination of photo-induced charge carriers in disordered semiconductors. In [34], the behavior of reaction front in $A + B \rightarrow C$ with subdiffusively moving reagents is studied. Consideration is based on a fractional-order reaction-diffusion equation containing motion and reaction terms affected by the subdiffusion. The continuous time random walk approach to the generalization of reaction-diffusion schemes is used in [11,35,36].

In the present work, kinetic models of aggregation and dissolution of clusters in disordered heterogeneous materials based on subdiffusive equations are studied within the generalization of the approach used in [24]. The decomposition of solid solutions and the formation of precipitates of the second phase are usually considered on the basis of the classical concepts of the diffusion of impurity atoms developed by Ham, Avraami, Aharon, Kotler, Lifshitz, and Slezov (see details in review [37]). These models actually do not admit the presence of inhomogeneities in alloys, which leads to an insufficiently complete reflection of transport phenomena occurring in real materials, for which the presence of disorder can lead to a significant change in the type of diffusion. The use of diffusion equations with fractional derivatives makes it possible to greatly simplify the consideration of such complex phenomena as diffusion along grain boundaries, dislocations, etc. In this work, the known models of the decomposition of supersaturated solid solutions are modified using various types of fractional differential operators. The difference from the works [23,24] lies in the generalization for the case of fractional operators with different kernels.

2. Generalized Fick's Law

In works [23,24], the kinetic models of aggregation and dissolution of clusters in disordered heterogeneous materials based on hereditary equations with fractional Riemann–Liouville derivatives are developed. A fractional-differential model of subdiffusion-assisted Ostwald ripening is proposed in [24]. The asymptotic behavior of time dependences of the supersaturation degree, the average cluster size, and other characteristics at the stages of subdiffusion-limited nucleation and coalescence are investigated. Here, we employ equations containing fractional-order operators with different memory kernels.

The hereditary Fokker–Planck equation for diffusion with memory effects has the form [38]

$$\frac{\partial}{\partial t} p(\mathbf{r}, t) = \frac{\partial}{\partial t} \int_0^t dt' \; K(t - t') \; \mathcal{L}_{\mathrm{FP}} \; p(\mathbf{r}, t'), \tag{1}$$

where $p(\mathbf{r}, t)$ is the impurity concentration, $\mathcal{L}_{\mathrm{FP}}$ is the following time independent operator

$$\mathcal{L}_{\mathrm{FP}} \; p(\mathbf{r}, t) = \frac{\partial}{\partial \mathbf{r}} \left[D(\mathbf{r}) \frac{\partial p(\mathbf{r}, t)}{\partial \mathbf{r}} - A(\mathbf{r}) p(\mathbf{r}, t) \right], \tag{2}$$

where $D(\mathbf{r})$ and $A(\mathbf{r})$ are the anomalous diffusion and advection coefficients, respectively.

Memory kernel $K(t)$ will be specified further, and it will be related to a certain type of fractional-order derivative. Rewrite this generalization in the form of the well-known fractional Fokker–Planck equation [39],

$$\frac{\partial}{\partial t} p(\mathbf{r}, t) = \hat{\mathcal{G}}_t \; \mathcal{L}_{\mathrm{FP}} \; p(\mathbf{r}, t), \tag{3}$$

where the integro-differential operator is

$$\hat{\mathcal{G}}_t p(\mathbf{r}, t) = \frac{\partial}{\partial t} \int_0^t K(t - t') \; p(\mathbf{r}, t') \; dt' \tag{4}$$

which corresponds to some fractional derivative of order $1 - \alpha$ with $\alpha \in (0, 1]$. Further we imply four different fractional operators $\hat{\mathcal{G}}_t$ (4). The choice of time-fractional derivatives depends on the physical mechanism of subdiffusion.

Particularly, the corresponding memory kernels can be as follows [38,40].

1. Riemann–Liouville (fractional) derivative of order $1-\alpha$ in (4) corresponds to

$$K(t) = \frac{t^{\alpha-1}}{\Gamma(\alpha)}, \qquad \tilde{K}(s) = s^{-\alpha}. \tag{5}$$

2. Truncated (tempered) fractional operator

$$K(t) = e^{-\gamma t} t^{\alpha-1} E_{\alpha,\alpha}\left((\gamma t)^{\alpha}\right), \qquad \tilde{K}(s) = \left[(s+\gamma)^{\alpha} - \gamma^{\alpha}\right]^{-1}. \tag{6}$$

3. Distributed-order fractional derivative

$$K(t) = \int_0^1 d\nu\ \rho(\nu) \frac{t^{\nu-1}}{\Gamma(\nu)}, \qquad \tilde{K}(s) = \int_0^1 d\nu\ \rho(\nu)\ s^{-\nu}. \tag{7}$$

The fractional orders $\alpha \in (0,1]$ and $\nu \in (0,1]$ correspond to the dispersion parameters of subdiffusion in disordered material, $\rho(\nu)$ is the distribution of fractional orders, γ is a truncation parameter, b_α is a constant, functions $E_\alpha(t)$ and $E_{\alpha,\beta}(t)$ correspond to the one- and two-parameter Mittag–Leffler functions, respectively.

In the next sections, numerical results are presented mainly for the first case of memory kernel, but analytical representations are derived for the general case.

From the continuity equation and Fokker–Planck Equation (3), one can derive the following expression for flux [38],

$$\mathbf{j}(\mathbf{r},t) = -\hat{\mathcal{G}}_t \left[D\frac{\partial p(\mathbf{r},t)}{\partial \mathbf{r}} - A(\mathbf{r})p(\mathbf{r},t)\right]. \tag{8}$$

This is the generalized Fractional Fick Law (FFL).

In our models of subdiffusion-assisted nucleation, we will use the solution of the fractional Fokker–Planck equation written in terms of subordination,

$$p(\mathbf{r},t) = \int_0^\infty \rho(\mathbf{r},\tau)\ q(\tau,t)\ d\tau, \tag{9}$$

where $\rho(\mathbf{r},t)$ is the solution of ordinary Fokker–Planck equation with the same initial condition. Variable τ is considered as the operation time. The Laplace transform of generalized subordinator $q(\tau,t)$ has the form [38]:

$$\tilde{q}(\tau,s) = \frac{1}{s\tilde{K}(s)} \exp\left(-\frac{\tau}{\tilde{K}(s)}\right).$$

The motivation for development of fractional models of nucleation in solid solutions is observation of scaling laws with power law exponents that are not consistent with predictions by ordinary Aaron–Kotler or Lifshitz–Slezov models, which predict exponents $1/2$ and $1/3$ for precipitate growth kinetics. Figure 1 demonstrates experimental time dependencies of average radius of Cu precipitates in α-iron. As we can see, values of the power law exponent determined by different authors lie between theoretical ones predicted by the Aaron–Kotler and Lifshitz–Slezov models.

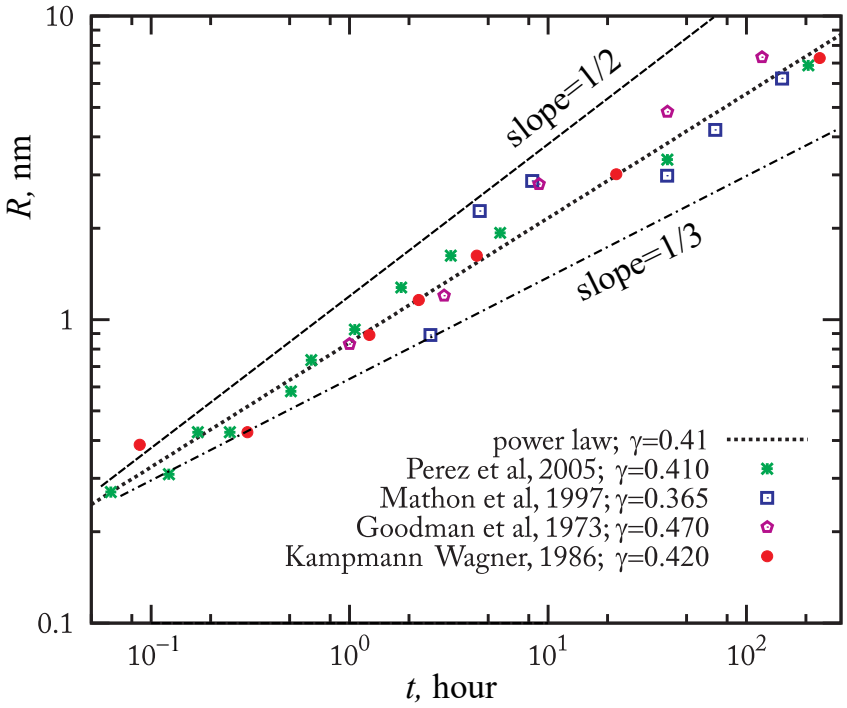


Figure 1. Experimental time dependencies of average radius of Cu precipitates in α-iron. Line is an approximation by a power law with exponent $\gamma = 0.41$. Slopes $1/2$ and $1/3$ in the log–log scale correspond to the case of normal diffusion-limited growth of isolated precipitate and the Ostwald ripening case, respectively.

3. Hereditary Generalization of Ham's Model

The Ham's theory [41] describes diffusion-limited growth of precipitates. The distribution of nucleation centers over the sample is assumed to be uniform. Usually, the cell containing the nucleus of the new phase in the center is selected. The size of this cell corresponds to average distance between neighboring clusters. The boundary conditions for this cell are [41]: $\rho(\mathbf{r}(S), t) = \rho_I$, $\vec{n}_T \nabla\rho(\mathbf{r}(T), t) = 0$. Further, we assume that the new phase precipitate has a spherical shape, and the diameter of the cluster is small compared to the distance between them.

Solving the boundary value problem under the indicated assumptions, it can be shown [23,41] that on the precipitate boundary

$$\nabla\rho(\mathbf{r}, t)|_{r=r_c} \approx \bar{\rho}(t)/r_c, \tag{10}$$

where $\bar{\rho}(t)$ is the average cell density of 'monomers' (unprecipitated particles).

After integrating the continuity equation, where $\mathbf{j}$ is determined according to the generalized Fick's law (8), over the volume of an equivalent sphere using Gauss theorem, we arrive at the following relation

$$\frac{dN}{dt} + \int_S D\,\hat{\mathcal{G}}_t\,\nabla\rho(\mathbf{r}, t)d\vec{S} = 0. \tag{11}$$

For the number N of 'free' (non-precipitated) particles in the cell, one can write:

$$N = \bar{\rho}(t)\frac{4}{3}\pi r_s^3. \tag{12}$$

The surface integral in (11) is reduced to multiplying the subintegral function by the sphere area. Using (12), we obtain:

$$\frac{4}{3}\pi r_s^3 \frac{d\bar{\rho}(t)}{dt} = -4\pi r_c^2 D\,\hat{\mathcal{G}}_t\,\nabla\rho(\mathbf{r}, t). \tag{13}$$

Substituting relation (10) into the right hand side of Equation (13), one can derive

$$r_s^3 \frac{d\bar{\rho}(t)}{dt} = -3r_c D\, \hat{\mathcal{G}}_t\, \bar{\rho}(t). \tag{14}$$

For the total number of particles (free and precipitated), the conservation law takes place:

$$\frac{4\pi}{3}\rho_c \left[r_c^3(t) - r_c^3(0)\right] = \frac{4\pi}{3} r_s^3 [\rho_0 - \bar{\rho}(t)]. \tag{15}$$

From relation (15), we have

$$r_c(t) = \left[r_c^3(0) + \frac{r_s^3}{\rho_c}(\rho_0 - \bar{\rho}(t))\right]^{1/3}. \tag{16}$$

Substituting this expression into (14), we obtain a fractional differential equation describing the kinetics of subdiffusion-assisted precipitation:

$$\frac{d\bar{\rho}(t)}{dt} = -\frac{3D}{r_s^3}\left[r_c^3(0) + \frac{r_s^3}{\rho_c}(\rho_0 - \bar{\rho}(t))\right]^{1/3} \hat{\mathcal{G}}_t\, \bar{\rho}(t). \tag{17}$$

Let us consider asymptotic relations following from Equation (17). For sufficiently small t, if $\bar{\rho}(t) \approx \rho(0)$, the equation becomes

$$\frac{d\bar{\rho}}{dt} = -\frac{3D\rho_0}{r_s^3}\left[r_c^3(0) + \frac{r_s^3}{\rho_c}(\rho_0 - \bar{\rho}(t))\right]^{1/3} \hat{\mathcal{G}}_t 1.$$

Using the variable separation method, one can derive the following

$$\bar{\rho}(t) \approx \rho_0 + \rho_c \frac{r_c^3(0)}{r_s^3} - \left(\frac{2D\rho_0}{r_s^2 \rho_c^{1/3}}\right)^{3/2} \left[\int_0^t \hat{\mathcal{G}}_\tau 1\, d\tau\right]^{3/2}. \tag{18}$$

Since t is small for the case $\frac{r_c^3(0)}{r_s^3} \ll \frac{\rho_0}{\rho_c}$, the latter expression can be replaced by another approximate relation:

$$\bar{\rho}(t) \approx \rho_0 \exp\left\{-\left(\frac{2D(\alpha)}{r_s^2}\right)^{3/2} \sqrt{\frac{\rho_0}{\rho_c}} \left[\int_0^t K(\tau) d\tau\right]^{3/2}\right\}. \tag{19}$$

This is a generalization of Ham's formula [41]. It should be noted that the diffusion coefficient D is also determined by a formula different from the classical one (for more details see [42]).

For large $t : \bar{\rho}(t) \ll \rho_0$, from Equation (17), we obtain

$$\frac{d\bar{\rho}(t)}{dt} = -\frac{3D}{r_s^3}\left[r_c^3(0) + \frac{r_s^3}{\rho_c}\rho_0\right] \hat{\mathcal{G}}_t \bar{\rho}(t). \tag{20}$$

Rewriting it with the use of the initial condition, we arrive at

$$\frac{d\bar{\rho}(t)}{dt} + K\hat{\mathcal{G}}_t\, \bar{\rho}(t) = \rho_0 \delta(t), \tag{21}$$

where

$$K = \frac{3D}{r_s^3}\left[r_c^3(0) + \frac{r_s^3}{\rho_c}\rho_0\right]^{1/3}. \tag{22}$$

Applying a fractional integral of order $1-\alpha$ to Equation (21), we obtain the following equation,

$$\frac{d^{\alpha}\bar{\rho}(t)}{dt^{\alpha}} + K\bar{\rho}(t) = \rho_0 \frac{t^{-\alpha}}{\Gamma(1-\alpha)}. \tag{23}$$

Solution for function $\bar{\rho}(t)$ is

$$\bar{\rho}(t) = \rho_0 \left[\frac{d}{dt} + K\hat{\mathcal{G}}_t\right]^{-1} \delta(t) \tag{24}$$

This dependence in asymptotic of large times is the power-law dependence with the exponent $-\alpha$:

$$\bar{\rho}(t) \sim \frac{\rho_0}{K} \frac{t^{-\alpha}}{\Gamma(1-\alpha)}, \quad t \gg K^{-1/\alpha}. \tag{25}$$

Results of simulation for time dependencies of supersaturation degree $\xi = \bar{\rho}(t)/\rho_0$ and precipitate radius R are presented in Figure 2. It can be seen that the analytical estimations agree with the numerical results.

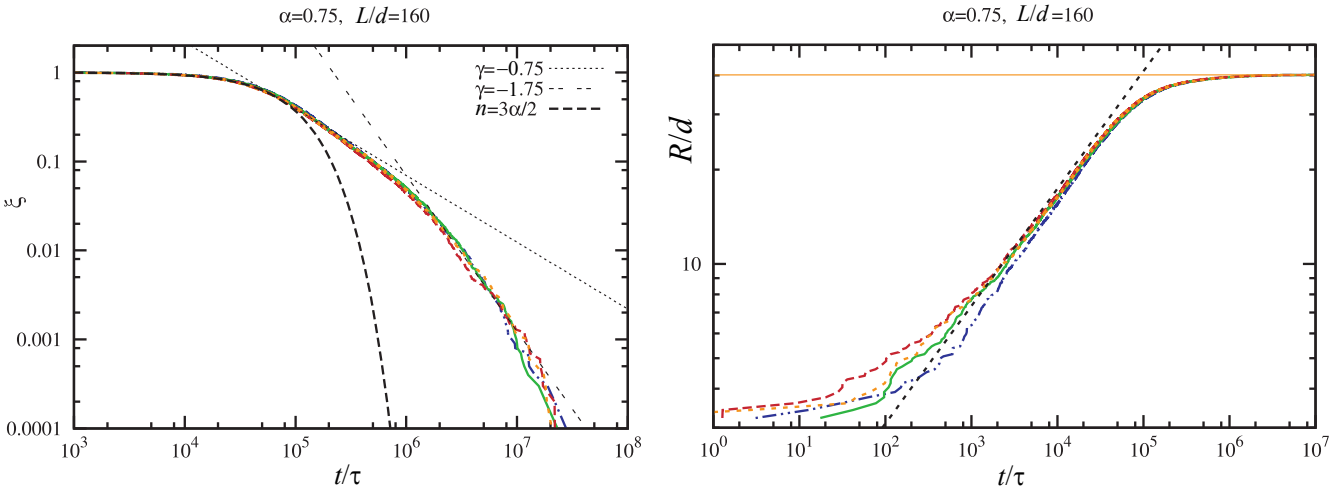


Figure 2. Time dependencies of supersaturation degree and precipitate radius in the fractional Ham model (with the Riemann–Liouville operator), $\alpha = 0.75$. Various colors of kinetic curves correspond to different realizations of the process.

Size change of the new phase precipitation can be obtained using relation (16), from which follows

$$r_c(t) = \left[r_c^3(0) + r_s^3 \frac{\rho_0 - \bar{\rho}(t)}{\rho_c}\right]^{1/3}. \tag{26}$$

At small times, we use (18) to obtain the expression

$$r_c(t) = \left[r_c^3(0) + R^3\right]^{1/3}, \tag{27}$$

where

$$R = \left(\frac{2D\rho_0}{r_s^2 \rho_c^{1/3}}\right)^{1/2} \left[\int_0^t \hat{\mathcal{G}}_\tau 1 \, d\tau\right]^{1/2}. \tag{28}$$

At large times from (21) follows

$$r_c(t) = \left[r_c^3(0) + r_s^3 \frac{\rho_0}{\rho_c}\left(1 - \left[\frac{d}{dt} + K\hat{\mathcal{G}}_t\right]^{-1} \delta(t)\right)\right]^{1/3}. \tag{29}$$

4. Hereditary Generalization of Aaron-Kotler's Model

In this section, using the generalized fractional diffusion equation, we consider the subdiffusion-assisted nucleation in an infinite matrix. Aaron and Kotler [43,44] solved the Stefan problem for the diffusion equation to describe the growth kinetics for an isolated spherical precipitate. The generalized model considers the modified boundary conditions for the case of fractional diffusion [24],

$$\rho(r = R, t) = \rho_I, \quad \rho(r \to \infty, t) = \rho_M, \quad t > 0; \tag{30}$$

$$(\rho_p - \rho_I)\frac{dR}{dt} = D\hat{\mathcal{G}}_t \left(\frac{\partial \rho}{\partial r}\right)_{r=R}. \tag{31}$$

The initial condition is assumed to be as follows

$$\rho(r, t = 0) = \rho_M, \quad r > R. \tag{32}$$

Here, ρ_p and ρ_M are the particle concentrations inside the precipitate and in the matrix far from it ($r \to \infty$), respectively. The equilibrium concentration near the particle surface is denoted as ρ_I.

In spherical coordinates, the equation of isotropic hereditary diffusion has the form

$$\frac{\partial \rho}{\partial t} = D\hat{\mathcal{G}}_t \left[\frac{\partial^2 \rho}{\partial r^2} + \frac{2}{r}\frac{\partial \rho}{\partial r}\right]. \tag{33}$$

Further we apply the "slowly moving boundary" approximation. It is valid, when $|\rho_M - \rho_I| \ll \rho_p - \rho_I$ [43]. Within the framework of this approximation, we solve the diffusion equation with the given stationary boundary condition at $R = \text{const}$. The precipitate radius $R(t)$ can be found after substituting this solution into relation (31).

Going over to the function $u(r, t) = r[\rho(r, t) - \rho_M]$, we obtain an equation, which after the Laplace transform in time becomes

$$\left[\tilde{K}(s)\right]^{-1} \tilde{u}(r, s) - \frac{\partial^2 \tilde{u}(r, s)}{\partial r^2} = 0.$$

The solution to this equation with specified boundary conditions has the form

$$\tilde{u}(r, s) = \frac{R(\rho_I - \rho_M)}{s} \exp\left(-(r - R)\left[\tilde{K}(s)\right]^{-1/2}\right).$$

Going over to the concentration, we get

$$\tilde{\rho}(r, s) = \frac{\rho_M}{s} + \frac{R(\rho_I - \rho_M)}{rs} \exp\left(-(r - R)\left[\tilde{K}(s)\right]^{-1/2}\right).$$

The substitution of this expression into the Laplace transform of Equation (31) gives the integral growth equation for spherical precipitates

$$R(t) = \frac{D}{\rho_p - \rho_I} \int_0^t K(t - \tau)\left(\frac{\partial \tilde{\rho}}{\partial r}\right)_{r=R} d\tau.$$

Particularly, for the tempered fractional diffusion defined by kernel (6), we have

$$\tilde{\rho}(r, s) = \frac{\rho_M}{s} + \frac{R(\rho_I - \rho_M)}{rs} \exp\left(-C^{-1/2}(r - R)\sqrt{(s + \gamma)^\alpha - \gamma^\alpha}\right).$$

For small times $t \ll \gamma^{-1}$ ($\gamma \ll s$), we deal with nucleation controlled by subdiffusion [24]

$$\tilde{\rho}(r,s) = \frac{\rho_M}{s} + \frac{\tilde{u}(r,s)}{r} = \frac{\rho_M}{s} + \frac{R(\rho_I - \rho_M)}{rs} \exp\left(-(r-R)\frac{s^{\alpha/2}}{\sqrt{C}}\right)$$

For large times $t \gg \gamma^{-1}$ ($s \ll \gamma$), the concentration takes the form of a classical Aaron–Kotler solution,

$$\tilde{\rho}(r,s) = \frac{\rho_M}{s} + \frac{\tilde{u}(r,s)}{r} = \frac{\rho_M}{s} + \frac{R(\rho_I - \rho_M)}{rs} \exp\left(-(r-R)\frac{s^{1/2}}{\sqrt{2C\sqrt{\gamma}}}\right)$$

The inverse Laplace transformation of these solutions leads to the following relations

$$\rho(r,t) = \rho_M + \frac{R(\rho_I - \rho_M)}{r} G_+^{(\alpha/2)}\left(t\left[\frac{r-R}{\sqrt{C}}\right]^{-2/\alpha}\right), \quad t \ll \gamma^{-1}, \tag{34}$$

$$\rho(r,t) = \rho_M + \frac{R(\rho_I - \rho_M)}{r} \operatorname{erfc}\left(\frac{r-R}{2\sqrt{Dt}}\right), \quad t \gg \gamma^{-1}. \tag{35}$$

Here, $G_+^{(\beta)}(t)$ is the cumulative function for one-sided Lévy stable law with exponent $\alpha/2$ [3], and $D = 2C\sqrt{\gamma}$ is a diffusion coefficient.

In a pure subdiffusive case (5) and size-independent ρ_I, we have the following fractional equation for the precipitate radius

$${}_0\mathrm{D}_t^{\alpha} R = C\frac{\mu}{R} + \sqrt{C}\,\frac{\mu t^{-\alpha/2}}{\Gamma(1-\alpha/2)}; \qquad \mu = \frac{\rho_M - \rho_I}{\rho_p - \rho_I}, \quad t \ll \gamma^{-1}, \tag{36}$$

$$\frac{dR}{dt} = D\frac{\mu}{R} + \sqrt{D}\,\frac{\mu t^{-1/2}}{\sqrt{\pi}}; \qquad t \gg \gamma^{-1}. \tag{37}$$

The scaling corresponding to (36) and (37) is as follows

$$R(t) = \begin{cases} \lambda_{\alpha/2} C^{1/2} t^{\alpha/2}, & t \ll \gamma^{-1}; \\ \lambda_1 (Dt)^{1/2}, & t \gg \gamma^{-1}. \end{cases} \tag{38}$$

5. Hereditary Generalization of Lifshitz-Slezov's Model

The late stage of precipitation, called coalescence or Ostwald ripening, is often characterized by the growth of large (supercritical) precipitates at the expense of small (subcritical) ones. During this process, the average cluster size growth, but the total area of the interfacial segregation decreases. This process plays a critical role in alloy hardening, emulsion stabilization, and nanoparticle synthesis. The coalescence controlled by normal diffusion is successfully described by the Lifshitz–Slezov–Wagner theory based on a system of equations, including the continuity equation in the space of sizes, the growth equation for individual particles, and the conservation law of total precipitate volume.

We consider the growth and dissolution limited by anomalous diffusion of impurities. Relation (31) can be written as

$$\frac{dR}{dt} = D\,\hat{\mathcal{G}}_t\left(\frac{\partial c}{\partial r}\right)_{r=R}. \tag{39}$$

Here, c is the solute concentration. The equilibrium distribution of the solution near the surface of a spherical precipitate of radius R is given by the following thermodynamic formula

$$c_I = c_I^{\infty} \exp\left(\frac{2\sigma w}{kTR}\right) \approx c_I^{\infty}\left(1 + \frac{2\sigma w}{kTR}\right).$$

Due to the low concentration of impurities at this stage, the impurity distribution near the precipitate can be described by the stationary distribution [45],

$$c(r,t) = c_M + (c_I - c_M)\frac{R}{r}.$$

The substitution $(\partial c/\partial r)_{r=R}$ into the continuity equation leads to equation

$$\frac{dR}{dt} = D\,{}_0\hat{\mathcal{G}}_t\,\frac{c_M - c_I}{R} = D\,\hat{\mathcal{G}}_t\left(\frac{c_M - c_I^\infty}{R} - \frac{2c_I^\infty\sigma w}{kTR^2}\right).$$

Rewriting the latter formula

$$\frac{dR}{dt} = \hat{\mathcal{G}}_t\left\{\frac{D}{R}\left(\Delta(t) - \frac{\vartheta}{R}\right)\right\}, \quad \Delta = c_M - c_I^\infty, \quad \vartheta = \frac{2c_I^\infty\sigma w}{kT},$$

Using the expression for critical radius $R_{\rm cr}(t) = \vartheta/\Delta(t)$, we arrive at

$$\frac{dR}{dt} = \hat{\mathcal{G}}_t\left\{\frac{R_{\rm cr}^3(0)}{R}\left(\frac{1}{R_{\rm cr}} - \frac{1}{R}\right)\right\}. \tag{40}$$

The continuity equation in the size space has the form

$$\frac{\partial f}{\partial t} + \frac{\partial}{\partial R}(v_R f) = 0, \quad N(t) = \int_0^\infty f(R,t)dR, \tag{41}$$

where $v_R = dR/dt$ is the growth rate and $N(t)$ is the number of precipitates per unit volume.

Equations (40) and (41) with the mass conservation law

$$\Delta(t) + \frac{4\pi}{3}\int_0^\infty R^3 f(R,t)dR = Q = \text{const} \tag{42}$$

provide the basis of the hereditary-diffusion coalescence model.

At a latter stage, as is follows from Equation (40), the nucleation scaling has the form

$$R(t) \propto \left(\int_0^t K(\tau)\,d\tau\right)^{1/3}.$$

For all fractional operators defined through Equation (4) and kernels (5)–(7), the correspondence principle is satisfied. When $\alpha \to 1$, we arrive at the well known scaling for normal coalescence $R(t) \propto t^{1/3}$.

6. Concluding Remark

The existing diffusion models of precipitation do not sufficiently reflect the properties of transport phenomena occurring in real materials, for which the presence of disorder leads to a significant change in the type of diffusion. The application of diffusion equations with fractional derivatives allows one to greatly simplify the consideration of such complex phenomena as diffusion in an inhomogeneous environment, along grain boundaries, dislocations, etc. Kinetic models of aggregation and dissolution of clusters in disordered heterogeneous materials based on hereditary equations with fractional derivatives are considered in the present paper. The asymptotic behavior of time dependencies of the supersaturation degree, the average cluster size, and other characteristics at the stages of subdiffusion-limited nucleation and coalescence are derived.

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